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Broadband integrated mid-infrared light sources as enabling technology for point-of-care mid-infrared spectroscopy

Alex Fuerbach
MACQUARIE UNIVERSITY

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"Broadband integrated mid-infrared light sources as enabling technology for point-of-care mid-infrared spectroscopy"

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Name of Principal Investigators (PI and Co-PIs): A/Prof. Alex Fuerbach and Prof. Mick Withford

- e-mail address: alex.fuerbach@mq.edu.au
- Institution: Macquarie University
- Mailing Address: Department of Physics and Astronomy, North Ryde, NSW 2109, Australia
- Phone: +61 2 9850 6145
- Fax: +61 2 9850 8115

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Abstract:

In this 1-year pilot project, we have investigated the possibilities to transform recently developed narrow-linewidth, continuous-wave (cw) ZBLAN chip lasers into robust and miniaturized broadband light sources for sensing applications. Utilizing nanomaterials-based saturable absorbers, we have demonstrated the possibility of passively q-switched as well as passively q-switched mode-locked (QML) operation of those chip lasers. Further, a novel nematic liquid crystal cell was successfully employed as an active q-switching element in the same type of chip lasers.

The short laser pulses that were generated in these experiments were finally launched into a highly nonlinear optical fiber for nonlinear frequency broadening and a maximum spectral bandwidth of close to 100 nm was obtained in the important 2 μm region of the spectrum. Numerical simulations reveal that further spectral broadening to a full mid-infrared supercontinuum is feasible.

Introduction:

Broadband light at mid-Infrared (IR) wavelengths (2 – 10 μm) has been suggested as a powerful and versatile diagnostic tool as this particular spectral region coincides with a vast number of highly specific molecular absorption lines. Potential applications include early cancer diagnostics, trace gas spectroscopy and environmental monitoring, areas that hold immense significance and importance.

However, laser source development at these wavelengths is still in its infancy, resulting in mid-IR spectroscopy being currently restricted to academic proof-of- principle experiments only. This project thus aimed to investigate options for the development a new class of broadband mid-IR lasers that are miniaturised, fully integrated, environmentally stable and robust. This would enable the integration of those sources into field transportable systems that could be used as true point-of-care diagnostics.

Experiments, Results and Discussion:

The main research directions that were pursued in this project can be divided into the development of pulsed waveguide chip lasers as well as into the investigation of subsequent spectral pulse broadening in a highly nonlinear optical fiber. The laser source development itself can further be subdivided into research into passively modulated laser systems that are based on low-dimensional nanomaterial saturable absorber materials and into actively modulated systems that are based on novel liquid crystal cell assemblies.

A. Passively modulated waveguide lasers

We have investigated the feasibility of integrating novel low-dimensional nanomaterials into the resonator of thulium-doped ZBLAN (ZrF_4-BaF_2-LaF_3-AlF_3-NaF) glass chip lasers with the aim of transforming these sources from continuous-wave (cw) lasers into pulsed lasers with correspondingly high peak power levels that would then allow them to be used as seeds lasers to excite nonlinear optical processes. We have concentrated on a range of different families of nanomaterials that all exhibit broadband saturable absorption which is a necessary prerequisite for their use as passive optical modulators in laser cavities.

We initially concentrated on optimizing the fabrication process for the individual absorbers. Chemical exfoliation followed by controlled spin coating was identified as the most reliable and reproducible method for fabricating saturable absorbers. We then set up an acousto-optically (AO) q-switched bulk laser with variable repetition rate to measure the nonlinear properties of the absorbers via the z-scan technique. Figure 1 below shows the performance characteristic of that probe laser as well as the result of a z-scan measurement performed on an absorber that consists of five spin coating layers of Black Phosphorus (BP). The measurement revealed a saturation intensity of the sample of 620 kW/cm^2, a modulation depth of 4.4% as well as non-saturable losses of 8.3%.

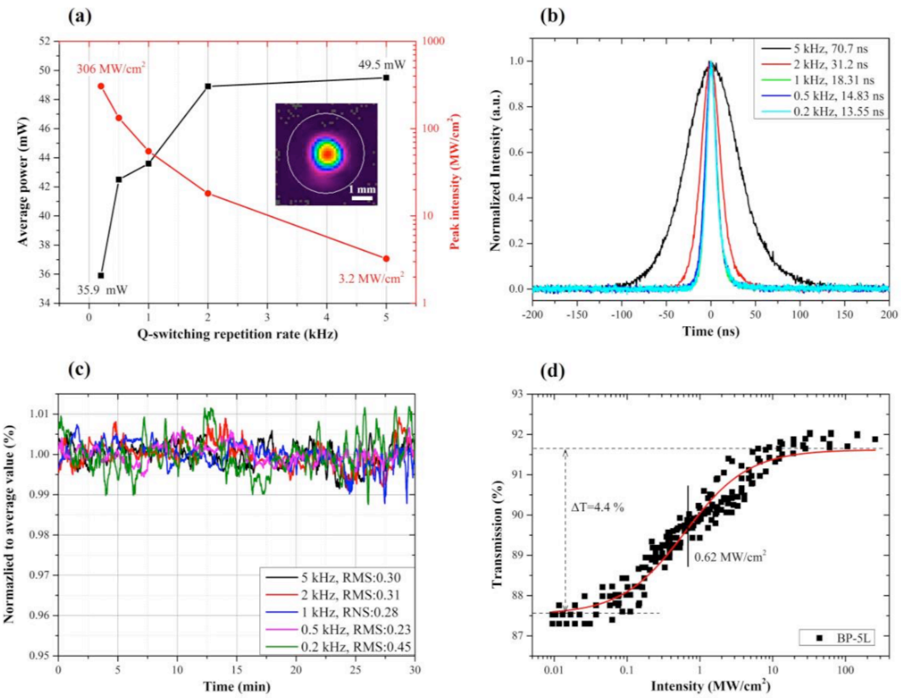

Figure 1. (a) Average output power and resulting peak intensity in the z- scan setup of the AO q-switched laser as a function of the laser repetition rate. The inset shows the output beam profile of the laser. (b) Oscilloscope trace of the laser pulses at different repetition rates. (c) Power stability of the laser at different repetition rates. (d) Measured nonlinear transmission of a BP saturable absorber with 5 spin-coating layers.

The materials that we included in our investigations were carbon nanotubes (CNTs), the topological insulator bismuth-telluride (Bi_2Te_3), graphene, black phosphorous (BP), indium tin oxide (ITO) as well as several transition metal dichalcogenides (TMDCs) like MoS_2, $MoSe_2$, WS_2 and WSe_2.
After individually optimising the fabrication process, we were able to successfully demonstrate q-switched as well as q-switched mode-locked (QML) operation of our waveguide lasers in the 2-micron range using any of the above-mentioned materials.

For pure q-switched operation, we have identified ITO as being the preferred material. It allowed us to achieve pulse durations of as short as 526 ns at repetition rates of 241 kHz and a maximum average power of about 100 mW, see figure 2 below.

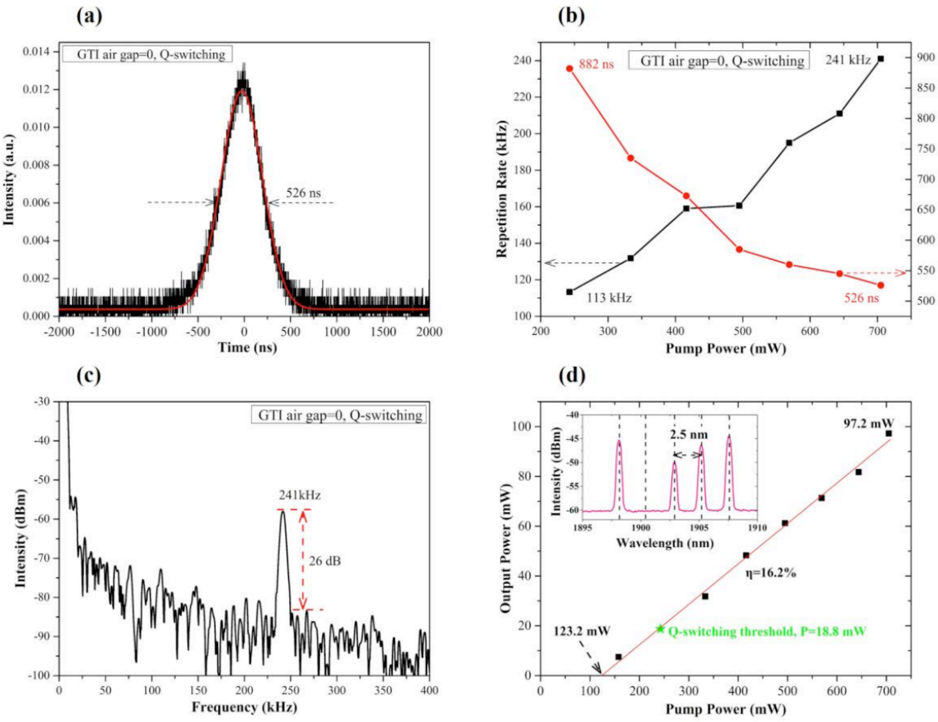

Figure 2. Performance of a passively Q-switched monolithic waveguide laser using ITO as saturable absorber. (a) Q-switched pulse at 705 mW pump power with a FWHM pulse duration of 526 ns. (b) Repetition rate and pulse duration as a function of pump power. (c) Radio frequency spectrum of the Q-switched laser at 705 mW pump power. (d) Output power as a function of pump power. The measured slope efficiency is 16.2% and the maximum output power is 97.2 mW. The insert shows the optical spectrum of the laser at maximum pump power.

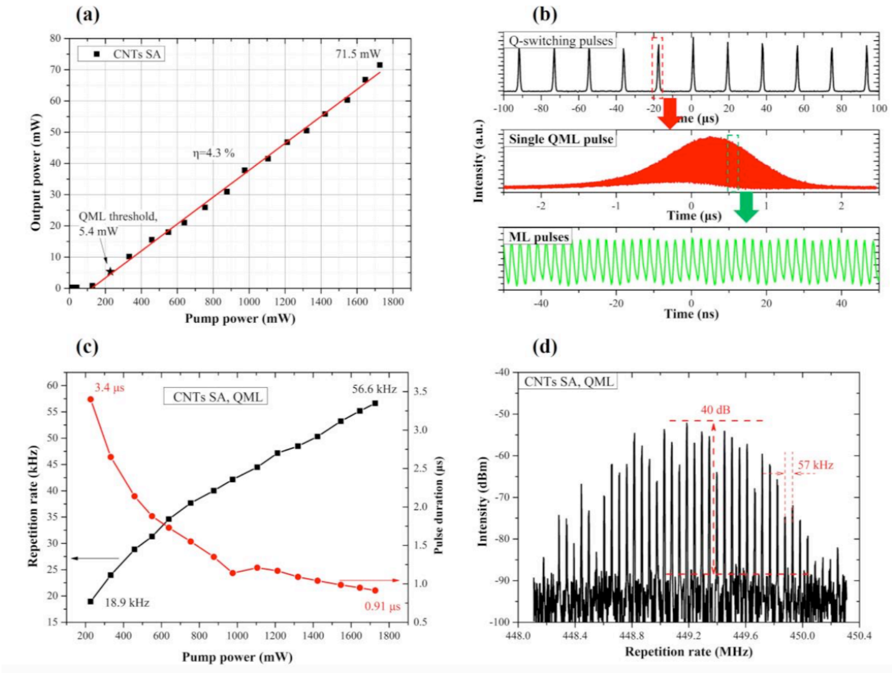

Figure 3. QML performance of the waveguide laser using the CNTs as saturable absorber. (a) Output power as a function of pump power. (b) Oscilloscope traces on different time scales. Top: Q-switched pulses train; middle: underlying mode-locked pulses in a single Q-switched pulse; bottom: mode-locked (ML) pulses train. (c) Q-switched repetition rate and pulse duration as function of pump power. (d) Radio frequency spectrum at maximum pump power.

For QML operation, we have identified CNTs as being the preferred material. Using CNT enabled us to generate low-noise q-switched pulses with underlying trains of ultrafast (mode-locked) pulses at a maximum average power of about 72 mW, see figure 3 for details.

B. Actively modulated waveguide lasers

We have previously demonstrated the possibility of using a novel nematic liquid crystal cell as an active q-switch in a near-infrared (i.e. at wavelength of around 1 µm) ytterbium-doped ZBLAN glass chip laser (Wieschendorf et al, Optics Express **25**, 1692, 2017). By optimising the composition of the liquid crystal cell, we were now able to translate our results into the 2 µm region. Most importantly, this shifts the output spectrum of the waveguide chip laser into the transparency window of highly nonlinear mid-infrared compatible glasses like chalcogenides (e.g. As_2S_3) and this is therefore paving the way for subsequent nonlinear spectral broadening. Using again a thulium-doped ZBLAN chip with femtosecond laser inscribed waveguides as the active medium, we successfully generated pulses with peak power levels approaching 500 W while at the same time keeping the average power low. This was possible as the active switch allows to operate the laser at relatively low repetition rates in the few-kHz range.
Figure 4 below shows a schematic of the setup used in our proof-of-principle experiments as well as the concept of a fully integrated waveguide laser.

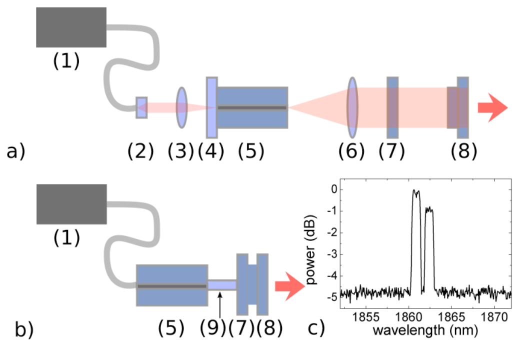

Figure 4. a) Schematic of the setup used in our experiments. (1) Laserdiode, (2) Collimator, (3) Focusing lens f=16 mm, (4) In-coupling mirror, (5) Thulium-doped ZBLAN waveguide chip, (6) Collimation lens f=50 mm, (7) Thin Film polarizer, (8) Liquid Crystal Cell, (9) GRIN lens. b) Fully monolithic setup. c) Spectrum of the laser.

C. Nonlinear spectral broadening

We finally launched the pulses that were generated by the actively modulated waveguide chip laser into a 1 m long section of a highly nonlinear As_2S_3 fiber which had a core diameter of 5 µm and a cladding diameter of 100 µm. As can be seen from figure 5 below, the extent of nonlinear spectral broadening reaches a width of almost 100 nm at the highest input peak power of about 450 W. Such a broad optical spectrum extending from about 1820 nm – 1920 nm would already be sufficient for spectroscopic analysis of a range of important molecules.

We have also performed numerical simulations of the pulse broadening process using the split-step Fourier method. Including the dispersive and nonlinear parameters of the used As_2S_3 fiber, those simulations show excellent agreement with the experimental results. Most importantly, these simulations also reveal that higher input peak power levels of about 2 kW would result in the formation of a broadband supercontinuum spanning from about 2 – 4 µm. We believe that by further optimizing our setup towards a fully integrated system and by thereby avoiding the excessive losses that are currently introduced by the used non-antireflection coated optics, a peak power level of > 2 kW should be entirely realistic to achieve. In addition, the materials that are currently used in the liquid crystal cell (the electrode material as well as the alignment layer material) could also be tailored

more specifically towards the mid-infrared spectral region of 2 microns. This again would result in an increased output peak power. It is also worth noting that the types of waveguides that are used in our experiments (depressed-cladding waveguides) offer the big advantage that very large mode-field diameters can be realised while still maintaining single transverse-mode operation of the laser. Thus, damage caused by high intracavity intensity levels can always be counteracted by an increase in mode-area.

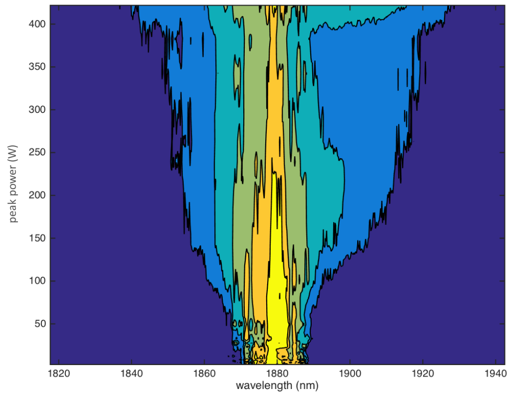

Figure 5. Spectral broadening as a function of input pulse peak power.

List of Publications and Significant Collaborations that resulted from your AOARD supported project:

Please note that a substantial amount of work for this project was carried out by Mr. Xiantao Jiang who submitted his PhD thesis in the first half of 2017. All three external examiners unanimously agreed that his work represents an excellent thesis. One examiner explicitly remarked that

"I am also a little surprised, given the breadth of work contained within the thesis that only one journal paper has resulted. I am assuming that the author has just concentrated on finishing this thesis and has not had time to write these papers. I sincerely hope that Xiantao Jiang will find the time to write these papers."

Indeed, we are currently in the process of writing those papers in which we will acknowledge the generous support that we have received from AOARD and that made this work possible.

<u>Papers published in peer-reviewed conference proceedings:</u>

1) C. Wieschendorf, J. Firth, S. Gross, M.J. Withford, D.J. Spence, F. Ladouceur, A. Fuerbach: "Mid-infrared monolithic pulsed waveguide laser that is actively Q-switched by a liquid crystal cell", The 24th Congress of the International Commission for Optics (ICO-24), Tokyo, Japan (2017)

2) A. Fuerbach, S. Gross, D. Little, A. Arriola, M. Ams, C. Wieschendorf, S. Antipov, X. Jiang, M. Withford: "Femtosecond laser direct-writing of integrated photonic devices", **invited talk**, 7th European Conference on Applications of Femtosecond Lasers in Materials Science (FemtoMat), Mauterndorf, Austria (2017)

3) C. Wieschendorf, X. Lei, J. Firth, L. Silvestri, S. Gross, F. Ladouceur, M. Withford, D.J. Spence, A. Fuerbach: "Compact actively Q-switched laser for sensing applications", 2nd International Conference of Fibre-optic and Photonic Sensors for Industrial and Safety Applications (OFSIS), Brisbane, Australia (2017)

4) C. Wieschendorf, J. Firth, L. Silvestri, S. Gross, F. Ladouceur, M.J. Withford, D.J. Spence, A. Fuerbach: "Novel liquid crystal cells for short-pulsed monolithic guided-wave laser sources", Conference on Lasers and Electro-Optics Europe (CLEO Europe), Munich, Germany (2017)

Interactions with industry that resulted from this work:

This work allowed us to strengthen an existing collaboration with the Australian Start-Up company Zedelef Pty. Ltd. We were initially working with Zedelef on the development of novel actively q-switched integrated waveguide chip lasers for the near-infrared region. The current grant enabled us to extend this collaborative work, and to include the 2-micron range and thus to investigate the potential use of those sources to drive a mid-infrared supercontinuum. This possibility, in turn, sparked the interest of the Lastek Group of Companies and we are currently in the process of finalising a research collaboration agreement between Macquarie University, Zedelef and Lastek to build a full prototype of the laser operating at 1 micron.